

10/717,628

(12) UK Patent Application (19) GB (11) 2 064 852 A

- (21) Application No 8037653
- (22) Date of filing 24 Nov 1980
- (30) Priority data
- (31) 101823
- (32) 10 Dec 1979
- (33) United States of America (US)
- (43) Application published 17 Jun 1981
- (51) INT CL³
G21F 9/28
- (52) Domestic classification
G6C FC
- (56) Documents cited
GB 2044982A
GB 1572867
GB 926401
- (58) Field of search
C7E
G6C
G6R
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(54) Decontaminating reagents for radioactive systems

(57) Surfaces contaminated with radioactive materials can be decontaminated by circulating an aqueous solution of decontaminating reagents which comprises organic acid decontaminating agents usually including oxalic acid, and formic acid, in contact therewith. The reagent preferably comprises both citric acid and EDTA in addition to oxalic and formic acids. It has been found that the efficacy of these organic acid decontaminating reagents can be prolonged under ionizing radiation by the inclusion of formic acid therein.

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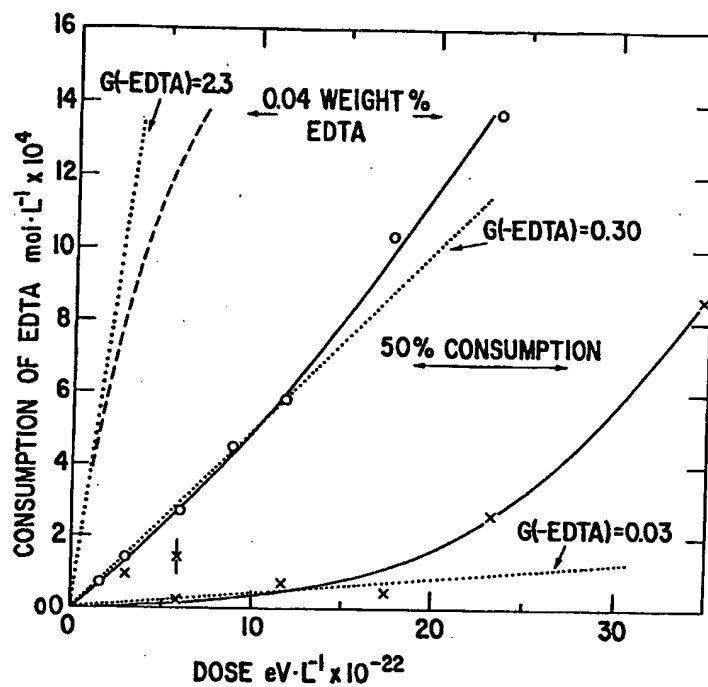


FIG. 1

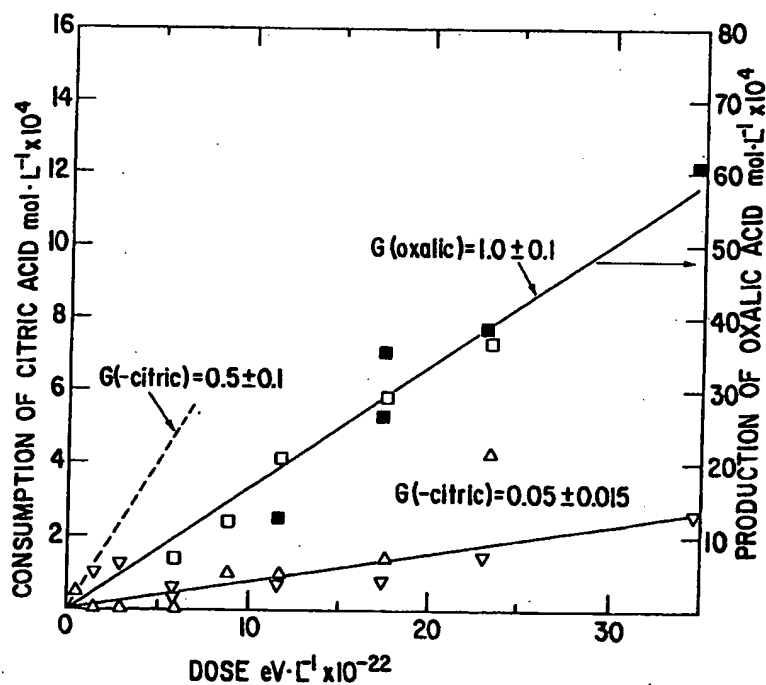
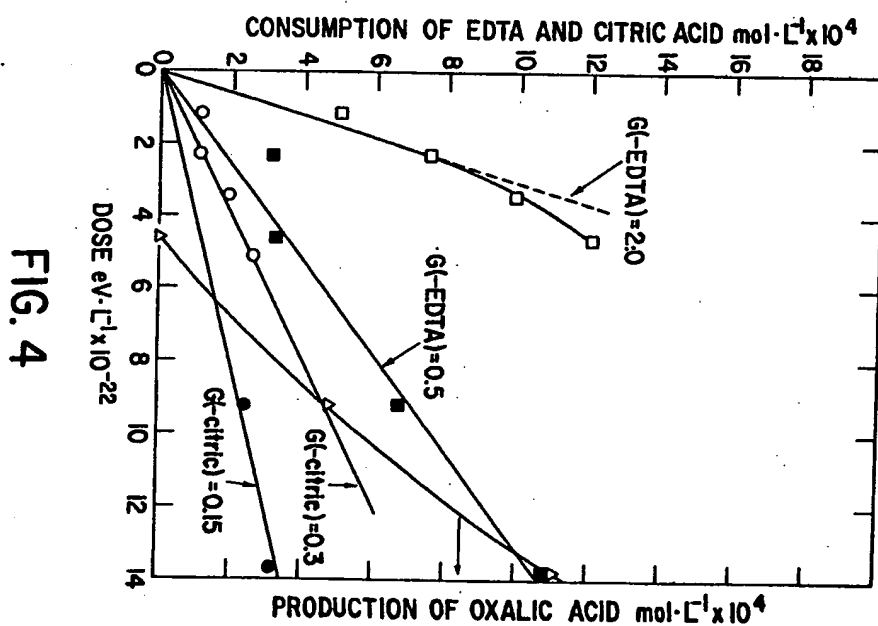
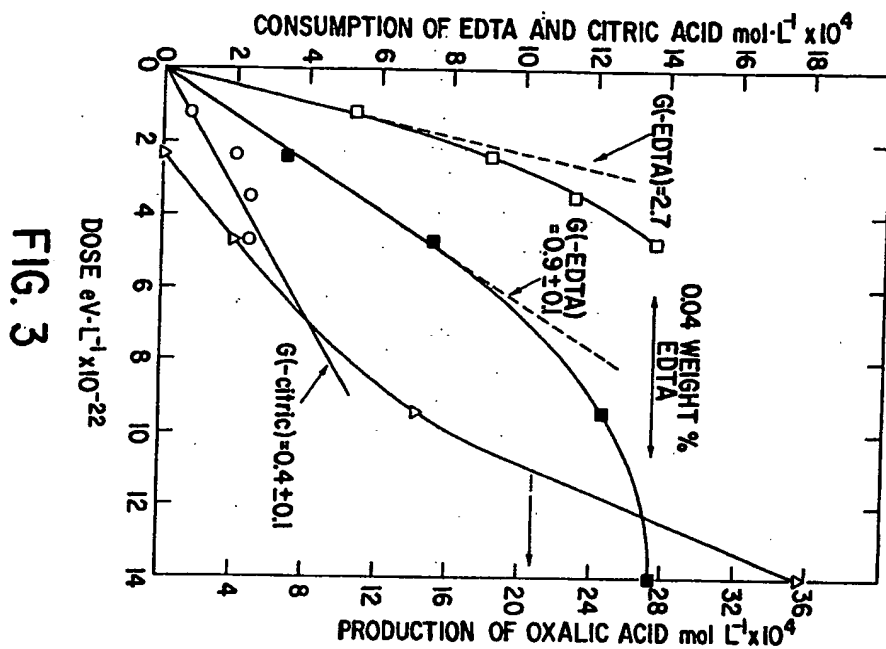
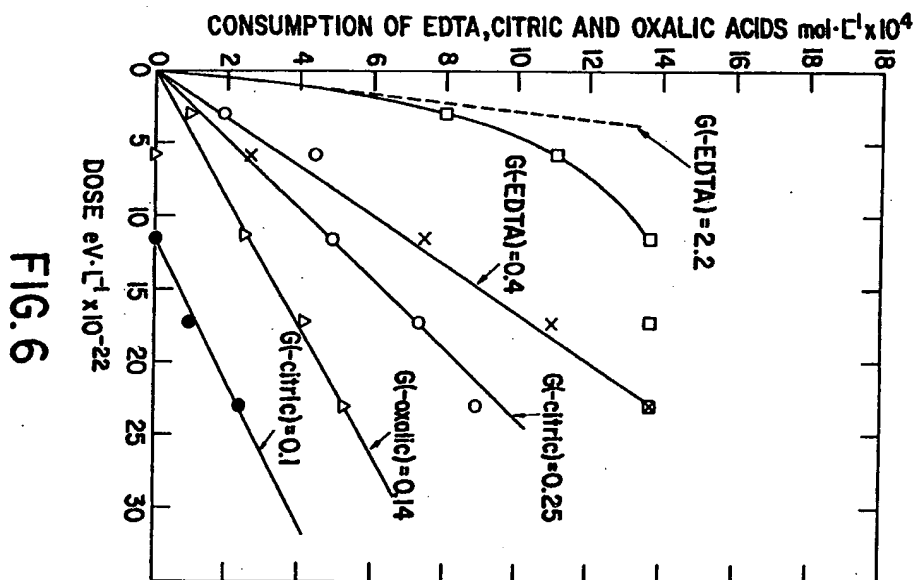
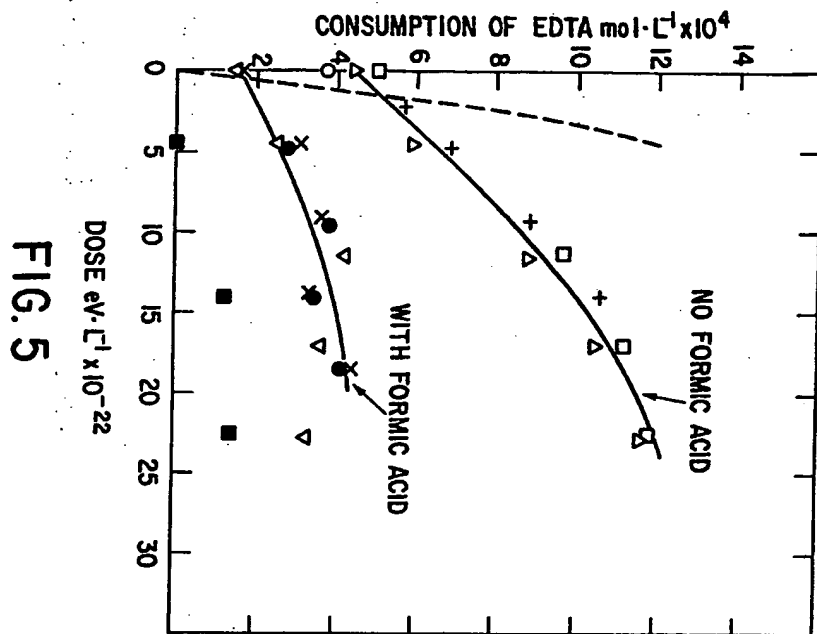


FIG. 2





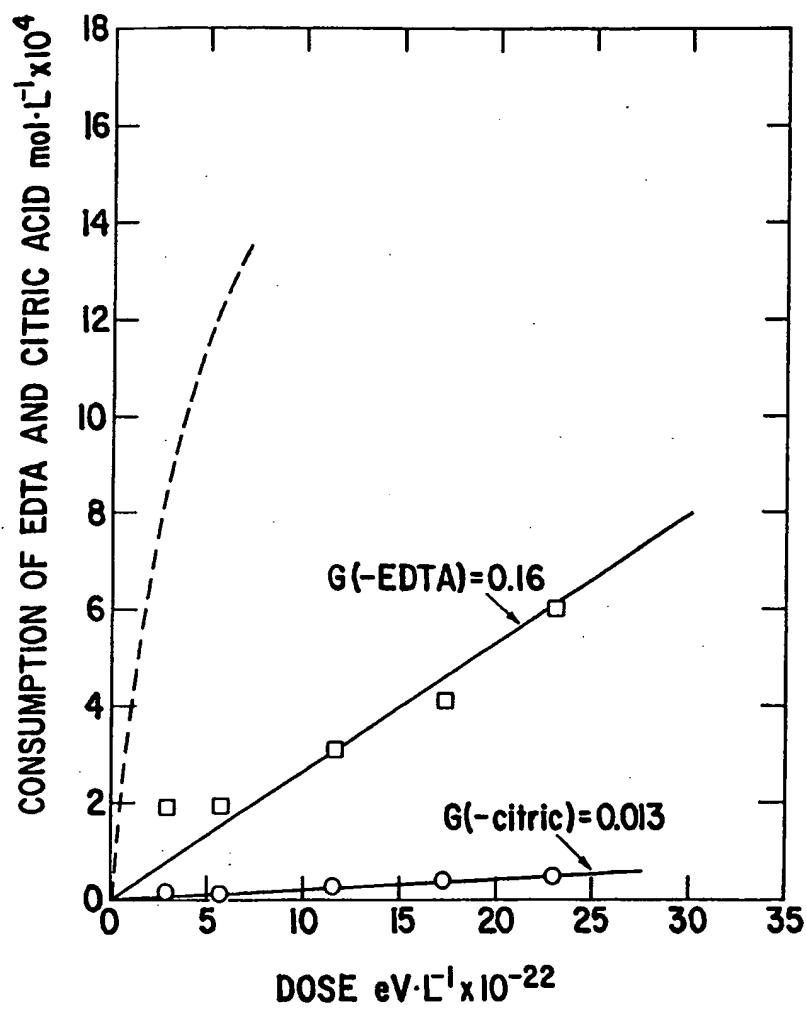


FIG. 7

SPECIFICATION

Decontaminating reagents for radioactive systems

5 This invention is directed to the decontamination of surfaces contaminated with radioactive materials, particularly coolant surfaces in nuclear reactors, by contact with circulating decontaminating solution. A novel decontaminating reagent mixture or reagent solution is provided having improved stability and prolonged efficacy. 5

Nuclear reactors having water coolant circuit surfaces on which radioactive deposits occur, have been treated with decontaminating reagent solutions to remove such surface contamination. Usually the treatment results in radioactive liquid wastes which are difficult and costly to dispose of. 10

United States Patent 3,272,738, September 13, 1966, discloses the precipitation of radioactive metal corrosion products from EDTA-hydrazine solutions by addition of sulfuric acid and ferrocyanide. United States Patent 3,873,362, March 25, 1975, similarly precipitates radioactive metal contaminants using permanganate, alkaline earth oxides, or alkaline earth hydroxides. United States Patent 4,162,229, July 24, 1979, utilizes a cerium IV salt as a decontaminating reagent. 15

Motojima et al in United States Patent 3,737,373, June 5, 1973, described the use of 0.1% deuterated oxalic acid in D₂O reactor coolant to dissolve contaminants. Irradiation of this oxalic acid-contaminant system caused decomposition of oxalates and precipitation of dissolved contaminants which were then recovered by filtration and/or ion exchange techniques. In this process, the deuterated oxalic acid is destroyed requiring additional deuterated acid to continue decontamination. The reactor must be cycled between cool, subcritical conditions and hot critical conditions in this Motojima et al process. 20

The "CAN-DECON" process has been developed by Atomic Energy of Canada Limited for decontamination of shutdown heavy water moderated and cooled reactors using dilute solutions minimizing corrosion and downgrading of the heavy water, giving a reduced volume of solid waste for disposal and relatively short decontamination time (see Canadian Patent 1,062,590, September 18, 1979, S.R. Hatcher et al). This method involves injecting selected acidic decontaminating reagents into circulating coolant to form a dilute reagent solution, circulating said solution to dissolve deposits and then to contact a cation exchange resin which collects the dissolved cations and regenerates the acidic reagents for recycle, and finally removing the reagents by contact with an anion exchanger to restore the coolant to its original condition (restoration of the coolant is particularly important with heavy water). Suitable acidic reagents used include ethylene-diaminetetraacetic acid (EDTA), oxalic acid, citric acid, nitrilotriacetic acid and thioglycolic acid. 25

The iron-EDTA complex has been proposed as an initial surface treating agent for steels in power plants (including nuclear) to create a good quality protective magnetite layer. M. Weber et al (Acta Chim. Acad. Sci. Hung. 97(3), 255-264, 1978) have studied the effect of ionizing radiation on the iron-EDTA complex + water surface-treating system and have found that the radiolytic decomposition of this ferric complex can be decreased by additions of methanol or formic acid. 30

Recently it has been found that these organic acid decontaminating reagent mixtures are subject to radiolysis in use leading to their decomposition over varying times with resulting loss of effectiveness for decontamination. Substantially complete consumption of EDTA was observed at radiation doses of about 0.8-1 Mrad. Citric acid used was also about 1/3 consumed at this dose, but the oxalic acid concentration remained stable. In the CANDU reactor decontamination process, it has been estimated that the average dose rate is about 0.3 Mrad per hour; leading to the disappearance of EDTA and substantial loss of effectiveness within about 3 hours. 40

In accordance with the present invention, it has now been found that multicomponent organic acid decontaminating reagents which may include oxalic acid and which are subject to radiolytic decomposition are improved by incorporating formic acid in sufficient amounts to prolong the efficacy of the mixture. 45

A preferred decontaminating reagent composition has been developed which comprises EDTA, citric acid, oxalic acid and sufficient formic acid to increase the stability and prolong the efficacy of the mixture on exposure to ionizing radiation. The formic acid has been found to decrease the decomposition of both EDTA and citric acid and to yield oxalic acid as a result of its own radiolysis. Thus formic acid causes minimal alteration of the reagent composition. This reagent composition is particularly advantageous for decontaminations where prolonged circulation through the contamination zone and cation exchange resins is carried out. 50

The invention includes, in the process for decontaminating surfaces having radioactive deposits in which a reagent mixture comprising organic acid decontaminating agents including oxalic acid is circulated over the contaminated surface, the improvement of initially incorporated formic acid in the mixture and maintaining the presence of formic acid by at least one further addition thereof. 55

In the accompanying drawings:

60 *Figure 1* is a graph showing radiolytic decompositions vs. radiation dose for EDTA in CAN-DECON solutions at pH 4.5 in the presence of sodium formate or formic acid. 60

Figure 2 is a similar graph showing consumption of citric acid and formation of oxalic acid vs. radiation dose in CAN-DECON solutions at pH 4.5 containing sodium formate or formic acid.

Figure 3 is a similar graph showing radiolytic decomposition of EDTA with sodium formate, and EDTA and citric acid without formate. The radiolytic production of oxalic acid in the presence of formate is also shown 65

(as in Figure 2).

Figure 4 is similar to Figure 3 and shows graphically the decomposition of EDTA and citric acid and production of oxalic acid in the presence of formic acid.

Figure 5 similarly depicts radiolytic decomposition of EDTA in the presence of dissolved iron with and without two concentrations of formic acid. The dashed line represents the control (no iron or formic acid).

Figure 6 similarly shows graphically the consumption of EDTA, citric acid and oxalic acid in the presence of boron and formate.

Figure 7 graphically shows the decomposition of EDTA and citric acid in the presence of ferric oxalate, ferrous sulphate and sodium formate with increasing radiation dose.

The presence of formic acid or formate ion is seen to decrease or retard the consumption (or decomposition) of the organic acids very considerably and thus to prolong the efficacy.

The amount of formic acid incorporated initially can vary considerably with even small amounts giving some benefit. Usually the amount of formic acid added is from about 1/4 to about twice the weight of the organic acid decontaminating agents subject to radiolytic decomposition. For the preferred EDTA, citric acid, oxalic acid, formic acid system, the proportions by wt. respectively may be 1:(0.6-1.2):(0.1-1.2):(0.6-3) but these are not critical. The relative proportions 1:1:1:1 are normally very suitable. In the relatively dilute CAN-DECON type of circulating solution, the concentrations of these same four components preferably would be approximately 0.03-0.05%; 0.02-0.04%; 0.01-0.04%; and 0.03-0.15% by wt. respectively. It is considered desirable to maintain the formic acid concentration at or above a minimum value of about 0.03% by wt. during the decontamination (by further additions thereof if necessary). Solution concentrations ranging up to saturation would be operative in situations other than CAN-DECON type decontaminations in CANDU reactors (such as in light-water-cooled reactors where the coolant may contain boron). Boron may also be present in heavy water coolant or moderator in some situations to control the neutron flux.

In these decontaminating reagent mixtures, on exposing to ionizing radiation, the formic acid radiolytic decomposition has been found to lead to the formation of some oxalic acid as well as H_2 and CO_2 . These gases can be removed by a degassing step as is frequently practised when upgrading the coolant for re-use. This additional oxalic acid is an added bonus. The original oxalic acid will itself be subject to radiolytic decomposition (to a lesser extent than some of the other acids) but usually there is a net gain in oxalic content on radiolysis of the mixture. Formic acid would not lead to any significant isotopic downgrading of the coolant in the case of heavy water coolant or moderator.

The organic acid decontaminating agents may comprise oxalic acid and usually one or more of EDTA, citric acid, nitrilotriacetic acid and thioglycolic acid. We have found mixtures of EDTA, citric acid and oxalic acid preferred in decontaminating heavy water reactor systems using CAN-DECON principles. The addition of hydrazine has been found advantageous for some applications. Other mixtures such as those including ascorbic acid or acetic acid or other acids mentioned above may be suitable for other systems, for example, light-water-cooled reactors.

In one way of carrying out the invention, we prefer to add the organic acid mixture to the coolant of a shutdown reactor and circulate the decontaminating reagents in solution contact with the surfaces being decontaminated until radioactive compounds are dissolved, the coolant solution subsequently being passed through a cation exchange resin column (in the H^+ or D^+ form) and through filters to remove radioactive cations and any suspended solids, and recirculated. Further addition(s) of formic acid may be desirable at an intermediate step. When the desired decontamination has been achieved, the solution is passed through both cation and anion exchange resins to remove dissolved metals and anions particularly the organic acid anions, thus restoring the coolant to normal. Where borate anions are present, some or all of the anion exchangers would be saturated with boric acid to ensure that sufficiently high borate ion concentration is maintained during each stage of the decontamination. A degassing operation to remove gaseous radiolysis products may be desirable before restoring the coolant to service.

The ion exchange resins are loaded as close to capacity as possible and usually the cation and mixed ion exchange resins (which are now highly radioactive) are disposed of as a solid waste material. Such a solid waste is more conveniently disposed of than a liquid waste, and with less environmental problems. Where heavy water coolant or moderator is utilized in the decontamination, the ion exchange resins would be converted to the D^+ and OD^- forms in order to avoid downgrading the deuterium content.

The following Examples are intended to be illustrative only. For test purposes, a standard decontaminating reagent consisted of EDTA, citric acid monohydrate and oxalic acid dihydrate in aqueous solution at respective concentrations of 0.04 (1.37); 0.03 (1.43); and 0.03 (2.38) weight % (the numbers in parentheses are in $mmol.L^{-1}$). For tests in the absence of dissolved metals the pH was adjusted to 4.5 (normal working range) by the addition of LiOH. All solutions were irradiated in 50 ml glass syringes using a Gammacell 220 (trademark) Co^{60} source at a nominal dose rate of $1.67 \times 10^{19} \text{ eV.L}^{-1}.\text{s}^{-1}$ (16 krad.min $^{-1}$). Solution temperatures in the Gammacell increased with absorbed dose but reached a steady state of 42°C after a dose of ~2 Mrad (2 hours). For test irradiation at 85°C the syringes were immersed in a thermostatically-controlled water bath, the dose rate falling to $1.3 \times 10^{19} \text{ eV.L}^{-1}.\text{s}^{-1}$ (12.5 krad.min $^{-1}$). The reagent acid components were separated and analyzed by gas chromatography after esterification with BF_3 in methanol. Dissolved iron concentrations (measured as Fe^{2+} and Fe^{3+}) were analyzed spectrophotometrically. Solutions containing dissolved iron were stored in the dark to prevent photolytic decomposition of the acids.

Example 1

The radiolytic decomposition (consumption) of EDTA at 0.04 wt% concentration in the standard decontaminating reagent solution was followed in the presence of 10 mmol.L⁻¹ sodium formate or 25 mmol.L⁻¹ formic acid at pH 4.5 and at Gammacell temperatures. At pH 4.5 the formic acid is present predominantly (85%) as the formate ion. Results are shown graphically in Figure 1. In Figure 1, the dashed line shows for comparison the consumption of EDTA in the absence of formic acid. 5

The addition of formic acid/formate clearly enhances the radiolytic stability of EDTA in these solutions compared to the control (no formate). The dose required to consume 50% of the EDTA increases from about 6 to 15 fold in solutions containing 10 to 25 mmol.L⁻¹ formic acid/formate. Initial disappearance yields correspond to G(-EDTA) = 0.30 and 0.03 molecules/100 eV respectively. The dotted lines on the graphs represent the initial slopes from which G(-EDTA) was calculated. These values can be compared with G(-EDTA) = 2.3 in the absence of formic acid/formate. 10

In Figure 2, the corresponding effects on citric acid and oxalic acid are shown under the same conditions, the dashed line being the consumption of citric acid in the absence of formate/formic acid. Again, relative to results obtained in the absence of formic acid/formate, the citric acid is protected about 10 fold with G(-citric) = 0.05 ± 0.015, whereas oxalic acid (right hand scale) is actually produced during radiolysis with a yield G(oxalic) = 1.0 ± 0.1. 15

Example 2

Radiolytic decomposition of EDTA and citric acid was followed in the test reagent solution at 85°C and pH 4.5. Results are shown on the graph in Figure 3. Oxalic acid concentration was unaffected at doses ≤ 4.7 × 10²² eV.L⁻¹ (≤ 0.75 Mrad). In the absence and presence of 10 mmol.L⁻¹ sodium formate, the left hand and center curves respectively show the effect on EDTA. The right hand scale shows a net production of oxalic acid under the conditions and in the presence of 10 mmol.L⁻¹ sodium formate. In this latter solution citric acid remained relatively unaffected at doses ≤ 14 × 10²² eV.L⁻¹ (≤ 2.2 Mrad). 20 25

Figure 4 shows the results of a similar experiment in the absence and presence of 10 mmol.L⁻¹ formic acid. Although the two graphs are not in perfect agreement, both EDTA and citric acid were protected and oxalic acid was produced at doses exceeding ~1 Mrad. In both graphs, Figures 3 and 4, at 85°C, EDTA was consumed at a rate about twice that observed at ≤ 42°C. 30

Example 3

The decomposition of EDTA at 85°C was followed in the same reagent solutions containing dissolved iron with and without formic acid. Solutions containing dissolved iron were prepared by heating de-aerated reagent solutions to 85°C in the presence of preoxidized carbon steel, Inconel 600 (trademark) and type 410 stainless steel up to pH 4.5. The oxidized metals had been prepared by autoclaving at 300°C in aqueous solutions adjusted to pH 10.5 with LiOH. This dissolution pretreatment would simulate an initial decontamination stage but in the absence of radiation. In the solutions containing 10 mmol.L⁻¹ formic acid, the total dissolved iron at pH 4.5 increased from about 260-290 ppm up to 480 ± 25 ppm largely as Fe²⁺ with the Fe³⁺ concentration remaining unchanged at 80 ± 10 ppm. Doubling the formic acid concentration to 20 mmol.L⁻¹ further increased the dissolved iron to about 725 ppm and the Fe³⁺ to 235 ± 35 ppm. The results are shown in Figure 5. The dashed line represents the EDTA results from Figure 4 without iron or formic acid. No decomposition of citric acid was observed at radiation doses ≤ 4 Mrad. 35 40

Before irradiation but after reaction with the preoxidized metal, the initial oxalic acid concentration had been decreased by about 75%, apparently due to the formation of a ferrous oxalate precipitate. Radiolysis with doses ≤ 5 Mrad did not decrease the oxalic acid concentration further and in some cases an increase was observed (the effect was not very reproducible). The initial exposure to preoxidized metal also decreased the EDTA by about 10% in the presence of 10 mmol.L⁻¹ formic acid, and by about 30% in its absence. A protective effect due to formic acid on the non-radiolytic decomposition of EDTA was achieved during initial metal dissolution, an extra advantage. Citric acid concentration remained stable. 45 50

From these tests with dissolved iron, it is indicated that the iron (predominantly Fe²⁺) also inhibits to some extent the radiolytic decomposition of EDTA and citric acid. This effect is further enhanced by formic acid but unlike the results presented in Figures 2-4, oxalic acid is not produced in comparable amounts. The overall effect observed on completion of this Example with formic acid and dissolved iron was enhanced protection for EDTA and citric acid with some reduction of the initial oxalic acid concentration due to precipitation. The formic acid was evidently also an aid to iron dissolution. 55

Example 4

In light water reactors boron is added to control the neutron flux for both regular operation and shutdown purposes. The effects of boron, if any, on the radiolysis of the test reagent mixture including formic acid are important with respect to the application of this invention to such reactor systems. 60

The decomposition of EDTA, citric acid and oxalic acid was followed as before but in the presence of

(a) 6000 ppm boron (added as H₃BO₃), and

(b) 2000 ppm boron and 10 mmol.L⁻¹ sodium formate.

The results are shown in Figure 6. The protective effect of the formate was comparable to that in the absence of boron (Figures 1 and 2). Although the initial consumption of oxalic acid (not shown) in the 2000 ppm 65

boron/formate solutions appeared to be increased over the 6000 ppm boron solutions, this effect was balanced by a compensating radiolytic production resulting in no net decomposition being detected after about 4 Mrad doses.

5 Example 5

Figure 7 shows the decomposition of EDTA and citric acid in the same starting solutions containing in addition 90 ppm Fe^{3+} as ferric oxalate, 45 ppm Fe^{2+} as ferrous sulfate and 10 mmol.L⁻¹ sodium formate. No net consumption or production of oxalic acid was detected at doses ≤ 4 Mrad. Thus the decontamination efficacy of the mixture would still be good after 4 Mrad radiolysis.

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Example 6

In order to determine whether the addition of formic acid had any effect on corrosion of carbon steel in the system and on decontamination of this steel, tests were run using the 0.1 wt.% standard test solution with and without 0.046 wt.% formic acid at 85°C. A test loop was used which included an ion exchange purification section. Corrosion runs were carried out on test coupons of carbon steel which had been pickled and degreased (and some prefilmed as in Example 3). Decontamination runs were carried out using test coupons of carbon steel which had been degreased and exposed to radiation in-reactor for eight weeks to render them radioactive. Also for decontamination runs, prefilmed carbon steel foil was added to the loop to cause approximate typical CAN-DECON iron concentrations in the solution. Before each test the loop was purged with N_2 and the water used was deoxygenated.

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The corrosion tests showed there was no significant difference in corrosion rate of the carbon steel due to the formic acid. Corrosion rates measured were from 0.12 to 0.18 $\mu\text{m}/\text{hour}$ for the pickled coupons, and from 0.15 to 0.25 $\mu\text{m}/\text{h}$ for the prefilmed surfaces. There was also no decrease in the decontamination and purification achieved with formic acid present. While these tests were conducted in the absence of radiolysis, under radiolytic conditions the formic acid would, as shown earlier, prolong the efficacy and performance of a given mixture.

25

Further tests with EDTA alone with formic acid showed a protective effect and prolonged efficacy of the mixture. After irradiation this mixture would include some oxalic acid, and would be useful for recirculation decontamination in some systems.

30

Figure 1 Caption

Radiolytic decomposition of EDTA at GAMMACELL temperatures in CAN-DECON solutions at pH 4.5:

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○ - containing 10 mmol.L⁻¹ sodium formate;
X - 25 mmol.L⁻¹ formic acid; dashed line shows, for comparison, the consumption of EDTA in the absence of formic acid. The dotted lines represent the initial slopes from which $G(-\text{EDTA})$ is calculated.

Figure 2 Caption

Radiolytic decomposition of citric acid and formation of oxalic acid at GAMMACELL temperatures in CAN-DECON solutions at pH 4.5:

40

Δ and ∇ , consumption of citric acid, and
 \square , \blacksquare , formation of oxalic acid (right hand scale) in solutions containing 10 and 25 mmol.L⁻¹ sodium formate and formic acid, respectively. The dashed line shows the consumption of citric acid in the absence of formate/formic acid.

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Figure 3 Caption

Radiolytic decomposition of CAN-DECON solutions at 85°C and pH 4.5: \square and \blacksquare , EDTA in the absence and presence of 10 mmol.L⁻¹ sodium formate, respectively; ○, citric acid (no formate); Δ , oxalic acid, production in the presence of formate.

50

Figure 4 Caption

Radiolytic decomposition of CAN-DECON solutions at 85°C and pH 4.5: \square , \blacksquare , EDTA and ○, ●, citric acid consumption in the absence and presence of 10 mmol.L⁻¹ formic acid, respectively. Δ , production of oxalic acid in solutions containing the formic acid.

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Figure 5 Caption

Radiolytic decomposition at 85°C of EDTA in simulated CAN-DECON solutions containing dissolved iron with and without formic acid.

5	Symbol	Fe ²⁺ (ppm)	Fe ³⁺	pH	formic acid mmol.L ⁻¹	5
	○	190	100	4.5	—	
	△	180	80	4.5	—	
10	□	195	90	4.5	—	10
	+	210	80	4.5-5.2	—	
	▽	385	90	4.5	10	
	X	390	70	4.5	10	
	●	380±20	110±25	4.3-4.4	10	
15	■	490±30	235±35	4.2	20	15

The dashed line represents the consumption of EDTA in the absence of dissolved iron or formic acid (from Figure 4).

Figure 6 Caption

Radiolytic decomposition of CAN-DECON solution in the presence of boron and formate ion: open symbols solutions containing 6000 ppm boron □, EDTA; ○, citric acid; △, oxalic acid; X and ●, EDTA and citric acid, respectively, in solutions containing 2000 ppm boron and 10 mmol.L⁻¹ sodium formate.

Figure 7 Caption

Radiolytic decomposition of CAN-DECON solutions at GAMMACELL temperatures in the presence of ferric oxalate (90 ppm Fe³⁺), ferrous sulfate (45 ppm Fe²⁺) and 10 mmol.L⁻¹ sodium formate. □, EDTA; ○, citric acid. The dashed line represents the decomposition of EDTA in the absence of additives.

CLAIMS

1. A decontamination reagent for decontaminating radioactive surfaces, comprising: an organic acid decontaminating agent subject to radiolytic decomposition or a mixture of two or more such agents, and formic acid in sufficient amount to prolong the efficacy of said decontaminating agent(s), said formic acid also yielding some oxalic acid on exposure of the mixture to ionizing radiation.
2. A reagent according to claim 1, wherein said decontaminating agent is or comprises ethylenediaminetetraacetic acid (EDTA).
3. A reagent according to claim 2, wherein said decontaminating agent comprises a mixture of
 - (a) EDTA;
 - (b) citric acid; and
 - (c) oxalic acid;
 and wherein the relative proportions by weight of (a), (b), (c) and formic acid are in the range 1:(0.6-1.2):(0.1-1.2):(0.6-3) respectively.
4. A reagent according to claim 3, wherein said relative proportions by weight are about 1:1:1:1 respectively.
5. A reagent according to any one of the preceding claims in the form of a solution of said decontaminating agent(s) and formic acid in water or heavy water.
6. A reagent according to claim 5 as dependent on claim 3 or 4, wherein the concentrations of (a), (b), (c) and formic acid in said solution are respectively 0.03-0.05%; 0.02-0.04%; 0.01-0.04%; and 0.03-0.15% by weight.
7. A reagent according to claim 5 or 6, wherein said solution also contains boron.
8. A process for the decontamination of radioactively contaminated surfaces which comprises contacting said surfaces in the presence of ionizing radiation with a reagent solution comprising oxidic acid and optionally one or more other organic acid decontaminating agents wherein there is initially present in the reagent solution formic acid.
9. A process according to claim 8, wherein the presence of formic acid is maintained in the reagent solution during the decontamination process by at least one further addition thereof.
10. A process according to claim 9, wherein the formic acid is maintained at a minimum concentration in said solution of about 0.03% by weight.
11. A process according to claim 8, 9 or 10, wherein the reagent solvent is light water or heavy water and the solution also includes boron.
12. A process according to claim 8, 9 or 10, wherein said surfaces are the cooling surfaces of a water-cooled nuclear reactor and wherein said reagent solution is formed *in situ* by adding formic acid and optionally one or more organic acid decontaminating agents including oxalic acid to the nuclear reactor coolant, and circulating the coolant over said surfaces.

13. A process according to any one of claims 8-12, wherein there is used as the decontaminating agent a mixture of EDTA, citric acid and oxalic acid.

14. A process according to claim 13, wherein the EDTA, the citric acid, the oxalic acid and formic acid are at least initially present in the relative proportions specified in claim 3 or 4.

Printed for Her Majesty's Stationary Office, by Croydon Printing Company Limited, Croydon, Surrey, 1981.
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